MOLECULAR ODER SYMMETRIES INVESTIGATED BY RESONANT (CARS, SRS) AND NON RESONANT (FWM, SHG) NONLINEAR POLARIZATION MICROSCOPY

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In the last few years non linear microscopy techniques have strongly evolved to become powerful imaging and spectroscopy tools that are nowadays ready to investigate the biological world. Indeed non linear microscopy allows spectroscopy and imaging of specific objects without the need of labeling or staining. This is the case of coherent Raman contrasts [1] that are resonant with a molecular vibrational level. In this case, the interaction of the incident fields with the sample induces a third-order nonlinear polarization, whose properties depend both on the vector nature and frequencies of the incident electromagnetic fields and the structure of the medium. We have recently used polarization resolved CARS to decipher the orientation of specific molecular bond in a crystal [2]. Indeed this study was possible because of the known structure of the targeted molecular bond. On the contrary, in bio-imaging, one deals with targeted molecular bond whose orientation and/or microscopic structure are unknown. In this framework it is very important to develop a general methodology to analyze polarization resolved signal in nonlinear microscopy.

In this work we investigate the potential of spherical basis to describe both the optical field and the induced nonlinear polarization. Theoretical investigations from the 70's [3] show that the complete accessible molecular function distribution can be recovered using polarization resolved methods and a complete spherical harmonic description of the susceptibility tensor, the electric fields and the molecular distribution function. We find that this model can be generalized for every *n*-wave mixing resonant and non-resonant processes. In this framework, this is our goal to investigate the impact of molecular symmetries on the recovered nonlinear polarization signals using non resonant (SHG, FWM) and resonant (CARS, SRS) contrast as generated in our microscope. We will discuss these concepts and give experimental illustrations in order to probe specific molecular organization in biological assemblies.

[1] such as coherent anti-Stokes Raman scattering (CARS) and stimulated Raman scattering (SRS)
[2] F. Munhoz, H. Rigneault, S. Brasselet, *Phys Rev Lett*, **105,** 2010, 123903
[3] P. D. Maker, *Phys Rev A,* **1,** 1970